

IN THE CLAIMS

Please amend the claims as indicated below, deleting text marked with strike through and adding the underline text. This set of claims replaces all previous claim sets.

1. (Currently Amended) Organic solutions containing metal(IV) salts and oxoacids of phosphorus from which, after evaporation of a solvent, insoluble compounds of general composition $M(IV)(O_3P-G)_{2-n}(O_3P-R^1-X)_n$ ~~can be~~ are obtained, where M(IV) is a tetravalent metal, -G is a generic inorganic or organic group, --R¹-- is an organic group, --X is an acid group and n is a coefficient ranging from 0 to 1.5.
2. (Previously Presented) The organic solutions of claim 1 wherein the tetravalent metal salt is an anion and is selected from the group consisting of carboxylates, chlorides and alkoxides.
3. (Previously Presented) The organic solutions of claim 1 wherein the tetravalent metal salt is selected from the group consisting of Zr, Ti, Sn and Ce or their mixture.
4. (Previously Presented) The organic solutions of claim 1 wherein the tetravalent salt is zirconyl propionate or chloride.
5. (Previously Presented) The organic solutions of claim 1 wherein the group -G is selected from the group consisting of acid groups --OH; --R²--SO₃H and --R²--PO₃H₂; and where --R²-- is an organic group with linear chain selected from the group consisting of --(CH₂)_m-- and --(CF₂)_m--.
6. (Previously Presented) The organic solutions of claim 1 wherein the group --R¹-- is an arylene group selected from the group consisting of --C₆H₄--; --C₆H₄--CH₂-- and --C₆H₄--CF₂--.

7. (Previously Presented) The organic solutions of claim 1 wherein the acid group --X selected from the group consisting of --SO₃H, --PO₃H₂ and --COOH.

8. (Previously Presented) The organic solutions of claim 1 wherein the organic solvent is a protonable solvent or solvents selected from the group consisting of N,N-dimethylformamide, N-methyl-2-pyrrolidone, dioxane, dimethylsulfoxide, acetamide, acetonitrile, alkanols and mixtures thereof.

9. (Currently Amended) The process for the insertion of nano-particles of tetravalent metal salts within the pores of polymeric or inorganic porous membranes comprising:

a) preparing the organic solution of claim 1 ~~which, at the same time, may also contain a polymer and/or an ionomer~~; b) impregnating the porous membrane with the solution; c) eliminating the solvent; d) repeating the steps b) and c) until a partial or complete pore filling is obtained; ~~and~~

wherein the tetravalent metal salts are phosphate-phosphonates; and

wherein the organic solution prepared in step a) optionally comprises a polymer and/or an ionomer.

10. (Cancelled)

11. (Currently Amended) A method of preparing proton conducting composite membranes, comprising:

a) preparing the organic solution of claim 1 ~~which, at the same time, may also contain a polymer and/or a proton conducting ionomer~~; b) impregnating a polymeric or inorganic porous membrane with the solution; c) eliminating the solvent; d) repeating steps b) and c) until a partial or complete pore filling is obtained; ~~and~~

wherein the tetravalent metal salts are phosphate-phosphonates; and

wherein the organic solution prepared in step a) optionally comprises a polymer and/or a proton conducting ionomer.

12. (Currently Amended) The proton conducting composite membranes of claim [[11]] 34 wherein the polymeric porous membrane is a polymer made of chemically and/or thermally stable polymers selected from the group consisting of, polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF), polyesters, polyethersulfones and fluoroelastomers.

13. (Currently Amended) The proton conducting composite membranes of claim [[11]] 34 wherein the pore dimensions of the porous membranes are in the range 0.02-20 μm and a porosity >10%.

14. (Currently Amended) The proton conducting composite membranes of claim [[11]] 34 wherein the phosphate-phosphonates selected from the group consisting of $\text{Zr}(\text{O}_3\text{P--CH}_2\text{--PO}_3\text{H}_2)_2$ and compounds of the series $\text{Zr}(\text{O}_3\text{P--OH})_{2-n}(\text{O}_3\text{P--C}_6\text{H}_4\text{--SO}_3\text{H})_n$, and $\text{Zr}(\text{O}_3\text{P--C}_6\text{H}_4\text{--SO}_3\text{H})_{2-n}(\text{O}_3\text{P--CH}_2\text{--PO}_3\text{H}_2)_n$, with n in the range 0.1-1.5.

15. (Previously Presented) A method of preparing conducting composite membranes, comprising:

a) preparing the organic solution of claim 1; b) impregnation of a porous ceramic membrane with the solution; c) eliminating the solvent; d) repeating steps b) and c) until a partial pore filling is obtained;

wherein the pores are partially filled with a tetravalent metal salt; and

wherein the membranes exhibit catalytic activity.

16. (Currently Amended) The ~~composite membranes~~ method of claim 15 wherein the tetravalent metal salts is a phosphate-phosphonate selected from the group consisting of $\text{Zr}(\text{O}_3\text{P--CH}_2\text{--PO}_3\text{H}_2)_2$ and compounds of the series $\text{Zr}(\text{O}_3\text{P--OH})_{2-n}(\text{O}_3\text{P--C}_6\text{H}_4\text{--SO}_3\text{H})_n$, and $\text{Zr}(\text{O}_3\text{P--C}_6\text{H}_4\text{--SO}_3\text{H})_{2-n}(\text{O}_3\text{P--CH}_2\text{--PO}_3\text{H}_2)_n$, with n in the range 0.1-1.5.

17. (Currently Amended) A method for the preparation of nano-polymers constituted by nano-particles of tetravalent metal salts, comprising,

a)preparing the organic solution of claim 1; ~~which, at the same time, may also contain an organic or organic polymer or polymers and thereby producing a matrix of organic or inorganic polymers soluble in the same solvents,~~ and b) elimination of the solvent ; ~~and~~

wherein the tetravalent metal salts are phosphate-phosphonates; and

wherein the organic solution prepared in step a) optionally comprises an organic or inorganic polymer or polymers thereby producing a matrix of organic or inorganic polymers soluble in the same solvents.

18. (Previously Presented) The method of claim 17 wherein the organic polymeric matrix is that of a proton conducting ionomer.

19. (Cancelled)

20. (Previously Presented) The method for preparing the nano-polymers and nano-ionomers of claim 18 wherein the elimination of the solvent is performed by evaporation or with a non-solvent of the polymer or ionomer.

21. (Previously Presented) Nano-polymers produced by the method of claim 17.

22. (Previously Presented) The nano-polymers of claim 21 wherein the matrix is a synthetic ionomer selected from the group consisting of perfluorosulphonic polymers, sulfonated polyetherketones (sPEK), sulfonated polyethersulfones and sulfonated polyvinylidenefluoride (sPVDF).

23. (Previously Presented) The nano-polymers of claim 21 wherein the nano-particles of tetravalent metal salts dispersed in the polymeric matrix exhibit proton conductivity $>10^{-2}$ S cm⁻¹ at 70°C. and 95% relative humidity.

24. (Previously Presented) The nano-polymers of claim 21 wherein the nano-particles of tetravalent metal salts are selected from the group consisting of $\text{Zr}(\text{O}_3\text{P--CH}_2\text{--PO}_3\text{H}_2)_2$ and compounds of the series $\text{Zr}(\text{O}_3\text{P--OH})_{2-n}(\text{O}_3\text{P--C}_6\text{H}_4\text{--SO}_3\text{H})_n$, and $\text{Zr}(\text{O}_3\text{P--C}_6\text{H}_4\text{--SO}_3\text{H})_{2-n}(\text{O}_3\text{P--CH}_2\text{--PO}_3\text{H}_2)_n$, with n in the range 0.1-1.5.

25. (Currently Amended) A method for the preparation of membranes constituted by nano-polymers, comprising:

a) preparing the organic solution of claim 1; ~~which, at the same time, may also contain an organic or organic polymer or polymers and thereby producing a matrix of organic or inorganic polymers soluble in the same solvents,~~ and b) eliminating of the solvent ; and

wherein the tetravalent metal salts are phosphate-phosphonates; and

wherein the organic solution prepared in step a) optionally comprises an organic or inorganic polymer or polymers thereby producing a matrix of organic or inorganic polymers soluble in the same solvents.

26. (Currently Amended) The method of claim ~~[[21]]~~ 25, wherein the membranes are nano-ionomeric proton conducting membranes.

27. (Currently Amended) A method for ~~an easy~~ insertion of a large variety of lamellar nano-particles of tetravalent metal salts in the membrane/electrode interfaces of PEM FCs, comprising preparing the organic solutions according to claim 1 and eliminating the solvent.

28. (Previously Presented) A method according to claim 27, further comprising the addition of ionomers and/or other proton conducting compounds soluble in the same solvents.

29. (Previously Presented) An electrochemical device comprising the nano-polymers of claim 26.
30. (Previously Presented) The electrochemical device of claim 29, wherein said device is designed for generating electrical energy from the oxidation of a fuel.
31. (Previously Presented) A fuel cell for electrical vehicles and/or for portable electrical devices comprising the electrochemical device of claim 29.
32. (Previously Presented) Indirect methanol and direct methanol fuel cells comprising the electrochemical device of claim 29.
33. (Previously Presented) A catalytic membrane reactors comprising the proton conducting composite membranes produced according to claim 11.
34. (New) Proton conducting composite membranes, wherein the porous polymeric or inorganic membranes are impregnated with the organic solution of claim 1, and wherein the solvent has been eliminated.